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THE STUDY OF THE INTERACTION OF INTENSE PICOSECOND LIGHT PULSE WITH MATERIALS (1) MEASUREMENT OF PICOSECOND PULSE WIDTH USING TWO-PHOTON CONDUCTIVITY IN GAAS. (2) THREE PHOTON CONDUCTIVITY IN CDS

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Maryland University
College Park, Maryland

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THE STUDY OF THE INTERACTION OF INTENSE PICOSECOND LIGHT PULSE WITH MATERIALS

A QUARTERLY TECHNICAL REPORT

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13. ABSTRACT

1. Measurement of picosecond pulse width using two-photon conductivity in GaAs:

   Two-photon conductivity effect in GaAs has been utilized to map the second order intensity correlation function of the picosecond laser pulse for the first time. Reproducible data were obtained. The contrast ratio is about 1.8. This is limited by the thickness of the sample. In principle this method can be extended to use very thin sample to obtained subpicosecond time resolution.

2. Three photon conductivity in CdS.

   Three photon induced photoconductivity effect has been observed in CdS polycrystalline sample with a mode-locked Nd: glass laser. The three photo absorption coefficient was measured to be 0.043 cm^3/GW^2.

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THE STUDY OF THE INTERACTION OF INTENSE PICOSECOND LIGHT PULSE WITH MATERIALS

I. Measurement of picosecond pulse width using two-photon conductivity in GaAs.

2. Three photon conductivity in CdS.

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I. Measurement of picosecond pulse width using two photon conductivity in GaAs:

The two photon conductivity in GaAs (Cr-doped high resistivity type) was investigated using a Nd: glass mode locked laser and was reported in the earlier publication[1]. The photo conductivity versus laser intensity in a log-log scale is shown in Fig. 1. At lower light intensities the photo conductivity shows a square law dependence on intensity and changes to a linear and sublimear dependence because of the thickness of the material and stimulated intra-band absorption. The square law dependence of the photo conductivity on intensity could in principle be utilized to map the 2nd order correlation function of the intensity of the laser pulse. This will give us a measure of the pulse width.

The experimental set up used in the preset experiment is shown in Fig. 2. ANd: glass laser was mode-locked using Kodak 9860 dye in dichloroethane solution. The cavity length was adjusted to give a mode locked pulse train width 4 nanoseconds period. The mode locked pulse train was partially reflected by a plane glass beam splitter onto an ITT photodiode the output of which was monitored on a 519 oscilloscope. Another beam splitter reflects part of a beam on a reference GaAs crystal through ND filters. The transmitted beam was attenuated by ND filters and then split into two equal perpendicular components by a 50% - 50% dielectric beam splitter. The two beams were then made to collide on a GaAs crystal using two 99% dielectric reflectors. Both the reference and signal GaAs crystals were cleaved from a Cr doped high resistivity GaAs wafer of thickness .35 mm. Indium solder was alloyed to the end faces of the crystals and ohmic contact was thus established. Both ends of the crystals were connected in

series with 1200Ω resistor through a 22.5 volts battery. The voltage developed across the resistance was monitored in a dual beam oscilloscope.

The mode locked train of pulses was monitored on a 519 oscilloscope. Almost 80% of the shots gave a single neat mode locked pulse train probably due to the contact dye cell used in the experiment. First, the slope two region of the two GaAs samples were confirmed. The ND-filters were adjusted to keep the GaAs samples well inside the slope two region.

The photoconductivity ΔG was calculated from the voltage "θ" across the resistor R (1200Ω).

$$\Delta G = \frac{\theta}{(v-\theta)} \times \frac{1}{R}$$

where $V = 22.5$ volts

$R = 1200\ \Omega$

In the slope two region, θ was of the order of .02 volts.

Since $\theta << V$, ΔG is proportional to V and so the voltage θ measured on the oscilloscope can be taken as a measure of the two photon conductivity (TPC). The crystal GaAs #1 monitors the photoconductivity produced by the overlap of the pulse with itself while the crystal GaAs #2 monitors the p.c. due to a single passage of the short pulses, thereby providing the usual reference signal. The TPC pattern is scanned by moving the crystal #1 along the direction M_1 to M_2 and plotting the ratio of the pulse height from Sample #2 as a function of distance. The result is shown in the figure 3. Ratios obtained have been normalized to 1 in the wings to conform with units defined in Ref (2) where T P C yield due to a single pass was assigned a value of 1/2. Only data points for which a single neat mode locked pulse train was monitored on the 519 scope

were plotted on the graph. Each data point was an average of 8 to 10 shots.

The contrast ratio for two photon conductivity is the same as for the TPF experiment. To effect a comparison, the two photon fluoresence as obtained by Dugnay et al [3, 4] for a modelocked and a free running laser is also drawn on the same figure 3.

In the TPC measurement, the points more or less follows the TPF curve for a mode-locked laser not that of a free-running or Q-switched laser as can be seen in Fig. 3. However, we get a contrast ratio of 1.75 only. This is because of the poor resolution of the crystal. GaAs used in thi s particular experiment is of thickness 0.35 mm and this correspondes to a resolution of ~ 4 p sec. $\left(\frac{t}{c} \times n,\ n = 3.4,\ c = 3 \times 10^{10}\ \mathrm{cm/sec}\right)$.

If we sample and integrate the Dugnay's curve over 4 p secs at various points of the curve, we would expect the TPC or TPF curve with ~ 1.8 contrast ratio.

Thus we show that the two photon conductivity can be used to measure the width of the picosecond pulses. Thin film crystals of GaAs or some other thin photoconductors (e.g Cd S_x -Se_{1-x}) are suggested for future experiments to measure picosecond pulses. The crystals like Cd S_x -Se_1 have lower two photon absorption cross section and hence will insure a better square law region over a wide range of intensities. This elimiates the narrow square law region of GaAs. To improve the resolution, it is required to perform the experiment with thin film crystals. In conclusion, we state that we measured the auto correlation of the pulse intensity using two photon conductivity in GaAs which yielded the same shape of the curve as got by Dugnay [3] but with a reduced contrast ration of 1.75 because of limited resolution of the detector.

II. Three photon conductivity in Cd S: -

The availability of powerful sources of optical radiation had made it possible to perform a variety of experiments involving many-photon transitions in the optical range of wavelengths. The two-photon absorption process had been observed in cadmium sulfide [5], in an investigation of the recombination radiation excited with light from a q-switched ruby laser. In a different investigation [6], the two-photon absorption coefficient was measured. B. M. Ashkinadye et al [7] reported an investigation of two-photon conductivity in Cd S at room temperature, excited with giant pulses from a ruby laser. However, the three photon process in Cd S with the use of Nd: glass laser was experimentally observed only recently. B. M. Ashkinadge et al [8] detected the luminescence emitted by by Cd S at 77° K excited by neodymium laser. Cd S has a forbidded band width of of 2.57 ev at 77° K, the light of a Nd: glass laser (1.17 ev) should produce the three quantum absorption. They detected the luminescence recombination radiation) at 5200 Å and found that the luminescent intensity depended on the excitation intensity as $I_{lum} = I_{excitation}^{3.4}$. They observed the luminescence when the excitation intensity was varied between 20 MW/cm^2 and 100 MW/cm^2. Since they were using a q-switched laser, they have to focus it to get higher intensities. With the availability of mode locked Nd:glass lasers, it is possible to produce upto a few gega watts/$_{cm}$^{2} without focussing. Further because of the high peak power, the absorption process can be easily observed in the case of picosecond pulse excitation. Arsene et al [9] used the three photon absorption process in Cd S to estimate indirectly the picosecond pulse width from a measurement of the decay of luminescence along the length of the crystal. We investigated the three-photon conductivity in Cd S at room temperature using a mode locked

Nd: glass laser. The photo conductive cell has a dark resistance of well over a meg ohm. Since the Cd S cell is sensitive to ordinary light, we enclosed the cell in a box and cut off the light from the flash lamp and other sources using optical filters which cut off light other than 1.06 micron laser source. The cell was connected to a 22.5 volts battery through a 127 Ω resistor. The voltage across the resistance was monitored and measured on a dual beam oscilloscope along with the laser pulse. The mode locking of the pulse from the laser was monitored on a 519 scope. The change in conductivity ΔG is estimated from the voltage measured (θ) across the resistance R (127 Ω).

$$\Delta G = \frac{\theta}{(V - \theta) R}, \quad V = 22.5 \text{ volts.}$$

The intensity of the excitation intensity was varied using calibrated neutral density filters.

The photo conductivity ΔG in millimhos versus relative laser intensity is shown in Fig. 4. A least square fit was made and the slope was found to be 2.9. This indicated a power law of $\sim I^3$, characteristics of a three photon process. The generation of non-equilibrium carriers can be due to absorption of non-phase matched second harmonic generation in Cd S. Since the second harmonic is of 5300 Å, it will be absorbed only as a result two photon process. Hence, photo conductivity due to such a process is of fourth order and will be very weak compared to a three photon process. Therefore the observed photo conductivity is due to three photon absorption in Cd S.

We estimated the three photon absorption coefficient from the measured photo conductivity. For this, we had to know the peak intensity of the pulse. We measured the total energy of the mode locked pulse using a calorimeter. Under similar conditions, the two photon flourescence was photographed using Rhodamine 6G. This gave a value of $\sim$ 3 psecs without measuring contrast

ratio. 1.4 ge ga watts/cm^2 of peak intensity gave a value of $\Delta G = 0.8$ milli mhos. The three photon conductivity ΔG_3 can be easily written for a transient process using Jick Yee's formaula[11] as

$$\Delta G_3 = \alpha \tau \frac{I_o}{3\hbar w}\left[1 - \frac{1}{(1+2K_3 I_o^2 L)^{1/2}}\right]$$

where

$\alpha \quad = \quad q\, \frac{a}{c}(\mu_e + \mu_h)$

$q \quad = \quad$ electronic charge

$\frac{a}{c} \quad = \quad$ 2 geometric factor

$\mu_e + \mu_h \quad = \quad$ mobility = 200 cm^2|volt-sec in Cds

$\tau \quad = \quad$ pulse width (3 psecs)

$I_o \quad = \quad$ 1.4 ge ga watts/cm^2

$L \quad = \quad$ thickness of the crystal $\approx$ 0.2mm

$\hbar w \quad = \quad$ 1.17 ev

$K_3 I_o^2 \quad = \quad$ three photon absorption coefficient (cm^{-1})

The thickness of the crystal was measured with a microscope and was found to be approximately 0.2 mms. From the expressions for ΔG_3, $K_3 I_o^2$ was estimated to be 0.167 am $^{-1}$. For I $\approx$ 1.4 gega watts/cm^2, K_3 was found to be 0.043 cm^3/Gw^2. In a recent paper Jick Yee[12] calculated the three photon absorption coefficient using Hartree Fock approximation with a three valence bands model for Cd S and his value of K_3 was 0.25 cm^3/ Gw^2. Aykinadze et al [13] reported a value of 2.5 am^3/G w^2 for K_3. Their experiment was done with a q-switched laser pulse and their intensity dependence was 3.4. Aresenev et al[9] estimated using mode locked pulses and they got a value of 0.02 cm^3 / Gw^2. Our experimental arrangement was similar to Aersenev's and is in the order of magnitude agreement with Jick Yee's theoretical value and Aresenev's experimental value gives one more evidence to the three photon generation process in Cd S.

B. M. Ashkinadrye et al [8] observed a power law of $I_o^{3.4}$ in their experimental investigation of the recombination radiation of the 5211 Å band from Cd S at 77° K after three photon absorption of Q-switch Nd: glass radiation. Since they conducted the experiment at 77° K, they observed a large number of excitons. The excitons formed another recombination channel. They explained a power law of $I^{2.6}$ in the case of luminescence of Cd S with two photons excitation [13] by assuming two recombination channels and the pumping of carriers from one recombination channel to another. This was supported by their observation of two recombination times exhibited by the decay of the photo current. The power law of $I^{3.4}$ could be similarly explained. However, in the case of three photon excitation at room temperature, the photo conductivity exhibited

a power law of $I^{3.0 \pm .2}$. Since we operated the crystal at room temperature, there are very little excitons formed and so only one recombination channel is present as evidenced by the single recombination time (long time constant of 20 μsecs) in the photo conductivity decay even at the highest light intensities. No fast recombination was observed corresponding to the deexcitation of the excitons. Further excitons don't contribute to photo conductivity. B. M. Ashkinadye et al[7] experimented on two photon conductivity in Cd S at room temperature excited with giant pulses from a ruby laser and observed a power law of $I^{2.0}$. Exploring these results, the slope of 3.0 ± .2 in the log-log plot of ΔG_3 versus I could be justified.

In conclusion, we observed the three photon conductivity in Cd S at room temperature using mode locked Nd:glass laser pulses and the three photon conductivity depended on excitation intensity as $I^{3.0 \pm .2}$. An order of magnitude estimate of the three photon absorption coefficient was found to be in fair agreement with the theoretically calculated values. The power law of I^3 could be utilized to measure third order intensity correlations of the picosecond pulses. Third order processes like this could be observed easily with the use of picosecond pulses because of higher peak intensity. When Q-switched pulses of the same envelope density as that of the mode locked pulse train were used to excite the CdS crystal, no observable signal was detected. This indicates the advantages of using picosecond pulses in investigating multi-photon processes in semi-conductors.

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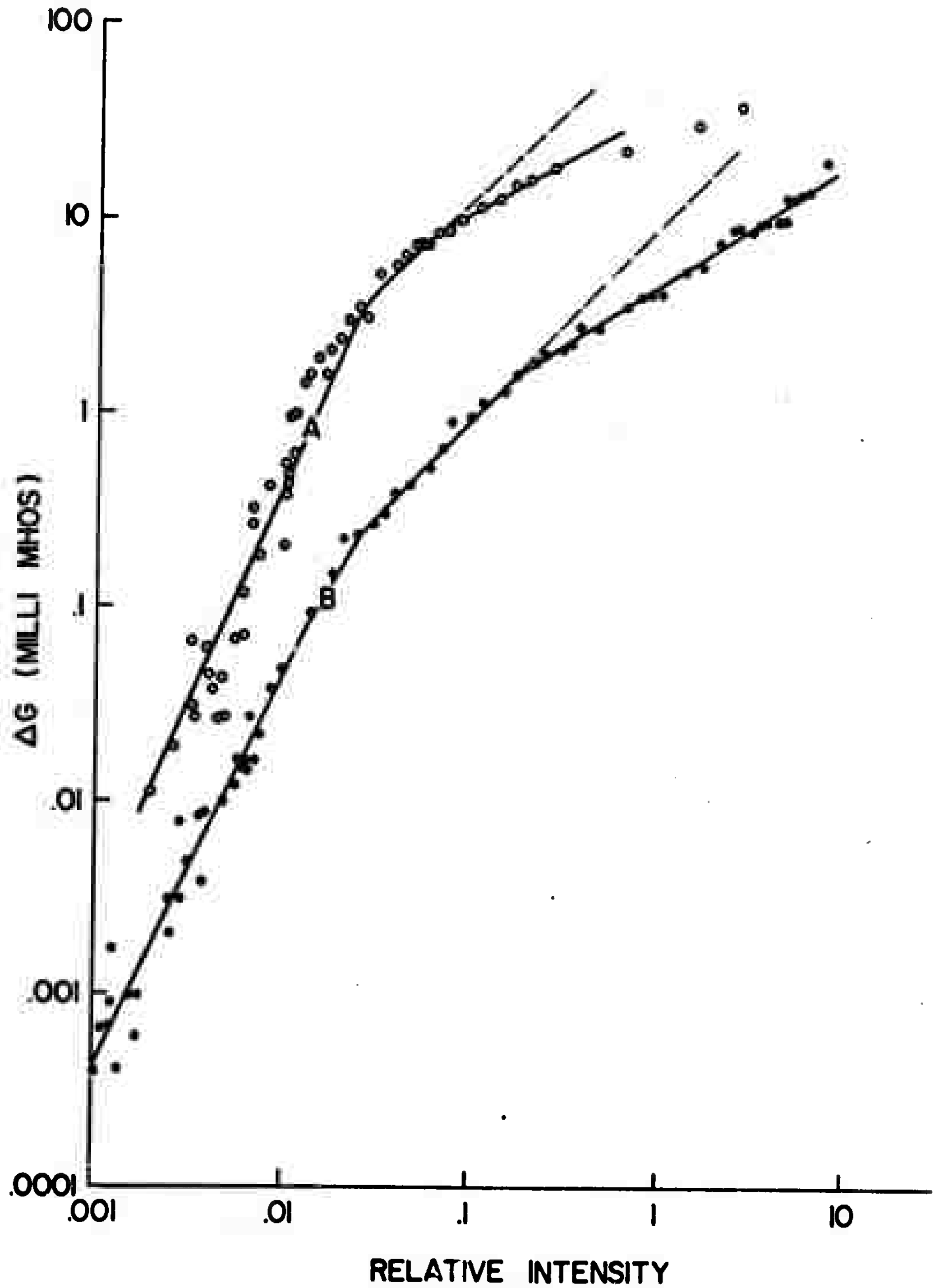
Figure Caption

Fig. 1 Two photon conductivity change in GaAs vs laser intensity with mode-locked pulse excitation.

Fig. 2 Experimental set-up for measurement of picosecond light pulses by using two-photon conductivity effect.

Fig. 3 Experimental data of second order intensity correlation curve as measured by two photon conductivity effect.

Fig. 4 Three photon conductivity change in polycrystalline CdS sample vs laser intensity.



MODE LOCKED PULSE EXCITATION
EXPERIMENTAL PHOTO CONDUCTIVITY

A n-TYPE O_2 DOPED GaAs (.028cm THICK)
B Cr-DOPED SEMI INSULATING GaAs (.033cm THICK)

LIFE TIME $\tau >> t_1$ (pulse width) $\approx$ 1 p sec

Fig. 1

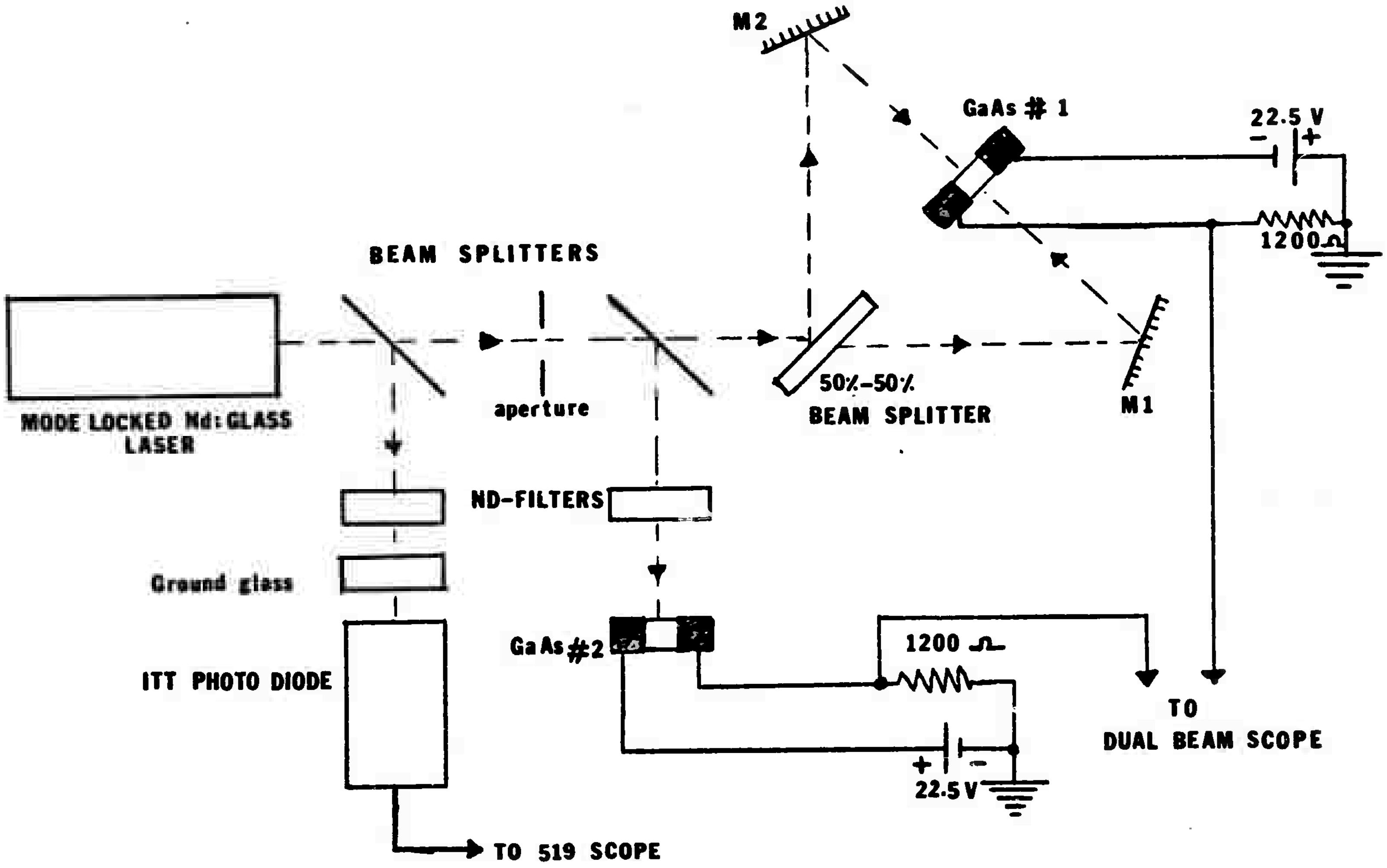

M2
GaAs # 1
22.5 V
1200 Ω
BEAM SPLITTERS
MODE LOCKED Nd:GLASS LASER
aperture
50%-50% BEAM SPLITTER
M1
ND-FILTERS
Ground glass
ITT PHOTO DIODE
GaAs #2
1200 Ω
22.5 V
TO DUAL BEAM SCOPE
TO 519 SCOPE

Fig. 2

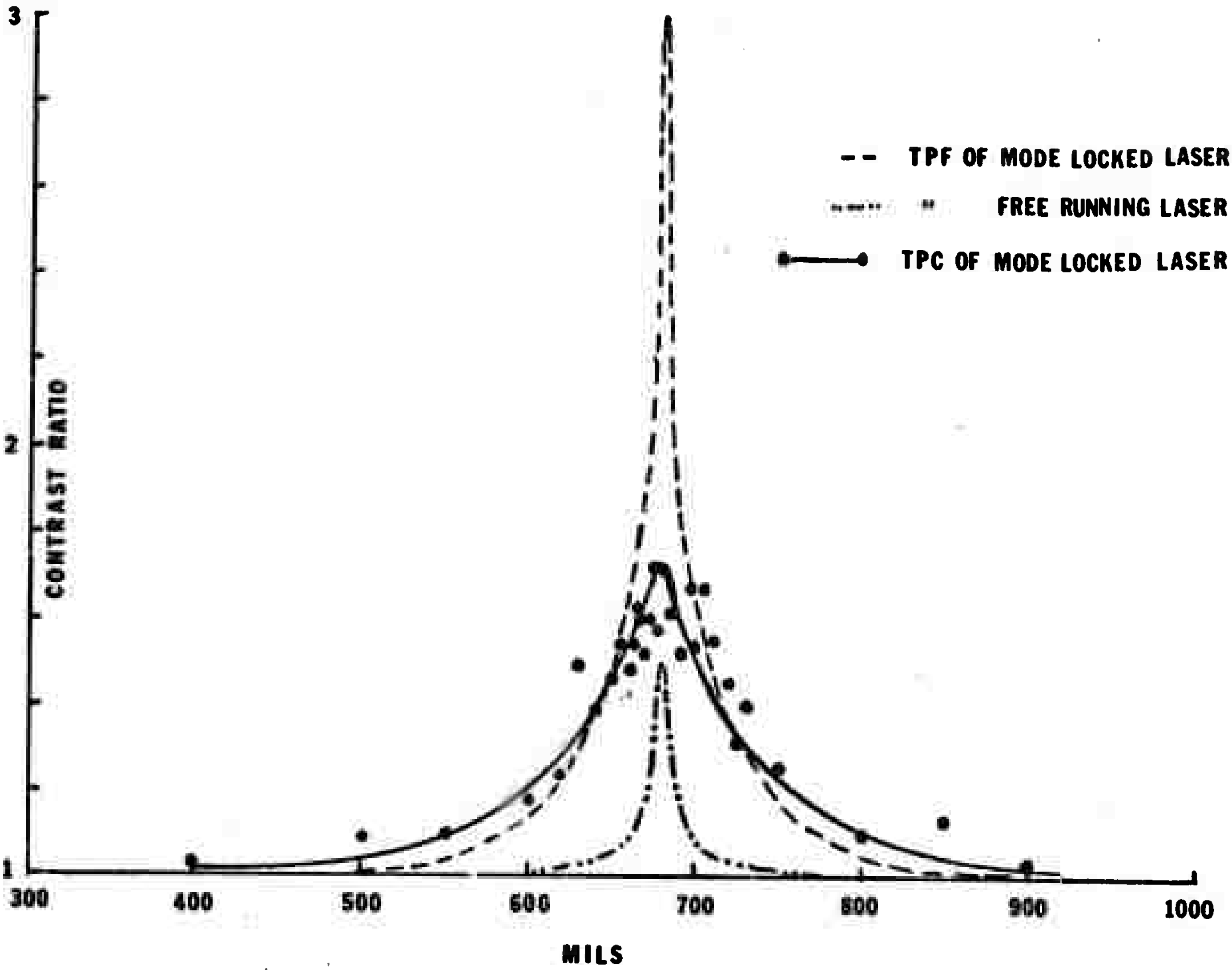
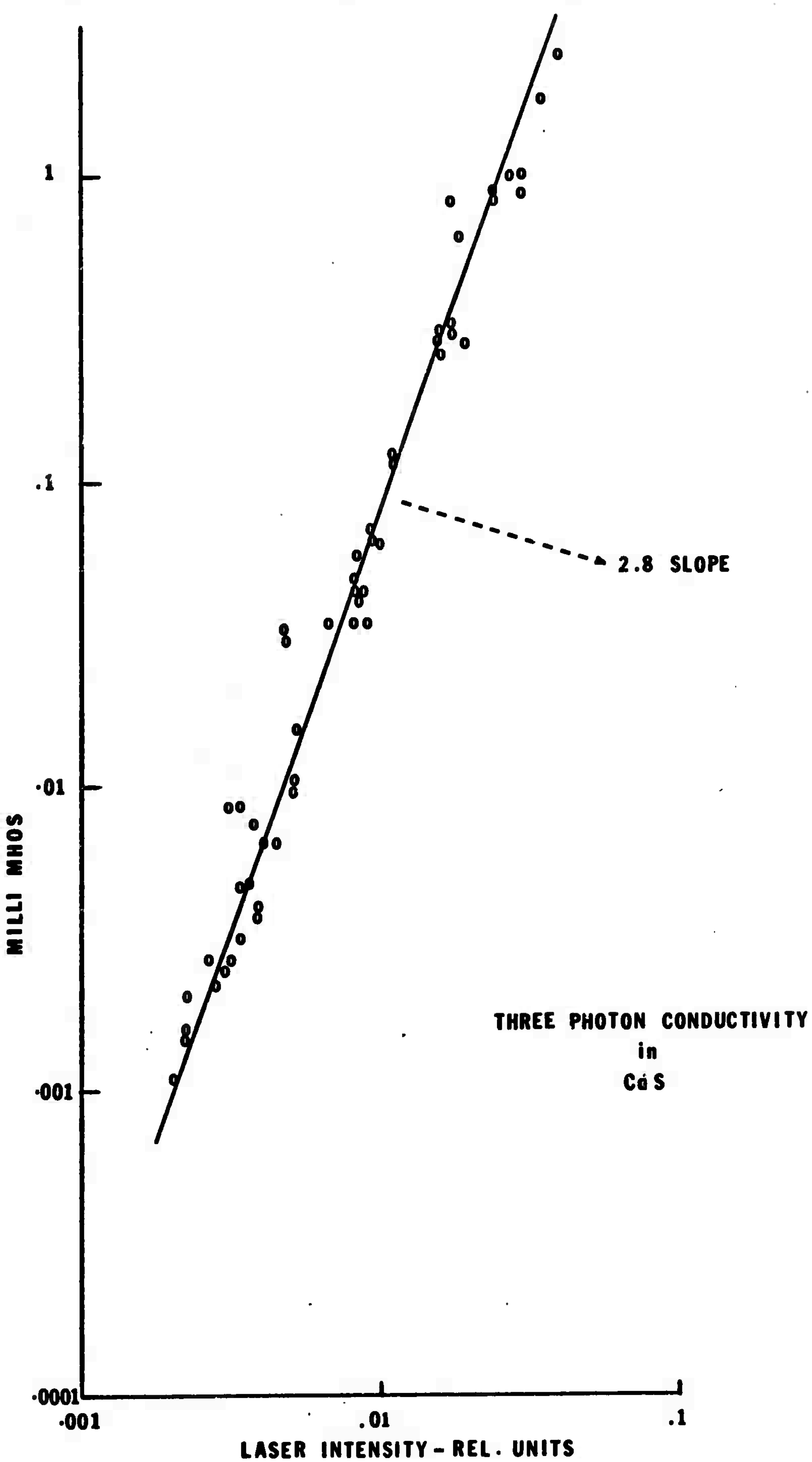

3
2
1
CONTRAST RATIO
TPF OF MODE LOCKED LASER
" FREE RUNNING LASER
TPC OF MODE LOCKED LASER
300
400
500
600
700
800
900
1000
MILS

Fig. 3


MILLI MHOS
1
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LASER INTENSITY - REL. UNITS
2.8 SLOPE
THREE PHOTON CONDUCTIVITY
in
Cd S

Fig. 4